

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: MICHEL, et al.

Docket: 2002DE142

Serial No. 10/533,999

Group Art Unit: 1796

Filed: 05/04/2005

Examiner: Khan, Amina S

For: Blue Dye With Particularly High Purity And Positive
Triboelectric Control Effect**DECLARATION UNDER 37 CFR § 1.132**

I, Dr. Hans-Tobias Macholdt, state that I am a resident of D-64297 Darmstadt-Eberstadt, Federal Republic of Germany; that I am a citizen of the Federal Republic of Germany; that I am a chemist having graduated at the University of Darmstadt, Federal Republic of Germany; that I am one of the inventors of U.S. Patent Application Serial No. 10/533,999; for "Blue dye with particularly high purity and positive triboelectric control effect"; that I am one of the inventors of US 5,061,585 and of US 6,168,895 cited as prior art against the beforementioned US Patent Application; that I consider myself qualified, by my knowledge of chemistry, and especially of triphenylmethane dyes, electrophotographic toners and developers and by my 20 years' experience in this field; that I can make the following observations and statements to wit:

This Declaration is to support by factual evidence the statements and observations of my January 14, 2009 declaration:

Aniline level according to US 3,652,602 (Schafer et al.)

While I showed in my previous Declaration an amount of 10,000 ppm aniline remaining in the final dyestuff if working according to Example 5 of this reference, there is an alternate purification method disclosed in this reference using a vacuum distillation of aniline (col. 3, lines 70-75). The distillation is described e.g. in Example 2 and was repeated as follows:

The complex-aluminum salt 4,4',4''-trichlorotriylchloride formed from 62.5 g of p-chlorobenzotrichloride, 200 g of chlorobenzene and 40 g of aluminum chloride was

converted by addition of 50 g of m-toluidine and by heating 3 hours at 130°C into the 4,4'-dichloro-4''-(m-methylphenylamino)-triphenylmethylchloride. To the hot melt was added 115 g of aniline and the temperature raised to 150-155°C. After 4 hours the melt was stirred into an equivalent amount of dilute sodium hydroxide solution to transform the dyestuff into the carbinol dissolved in aniline and for dissolving the aluminium chloride in the form of the aluminate. Then the mixture was boiled for 10 minutes and the alkaline solution separated off. The aniline was distilled off from the carbinol solution in vacuo at 160-170°C, 1 mm Hg, whereby the carbinol base was converted into an anhydro-base. Then, the anhydro-base was converted into the sulfate salt of formula (1) by adding an equivalent amount of sulfuric acid. The dyestuff sulfate was filtered off with suction, washed to neutral and dried in vacuo, a dark violet powder was obtained. Yield about 120 g.

Analytical HPLC method for determining aniline level:

Column: RP-select B;

Eluent: methanol/water, beginning with 30% methanol and 70% water, then graduating over the course of 20 min to 50% methanol; then to 100% methanol over the course of 10 min, then remaining constant at 100% methanol for 10 min;

Temperature: 40°C;

Flow rate: 0.2 ml/min.

Result:

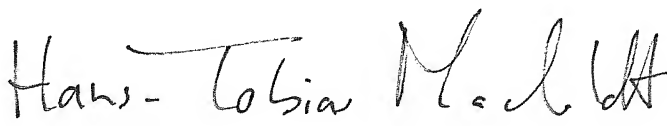
15000 ppm of aniline.

Evaluation of the results:

This experiment clearly demonstrates that not before the invention of U.S. Patent Application Serial No. 10/533,999 existed triphenylmethane colorants having aniline levels below 2,000 ppm.

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Frankfurt on the Main
June ³....., 2009

A handwritten signature in cursive script, reading "Hans-Tobias Macholdt". The signature is written in dark ink on a white background. Below the signature is a horizontal dotted line.

(Hans-Tobias Macholdt)